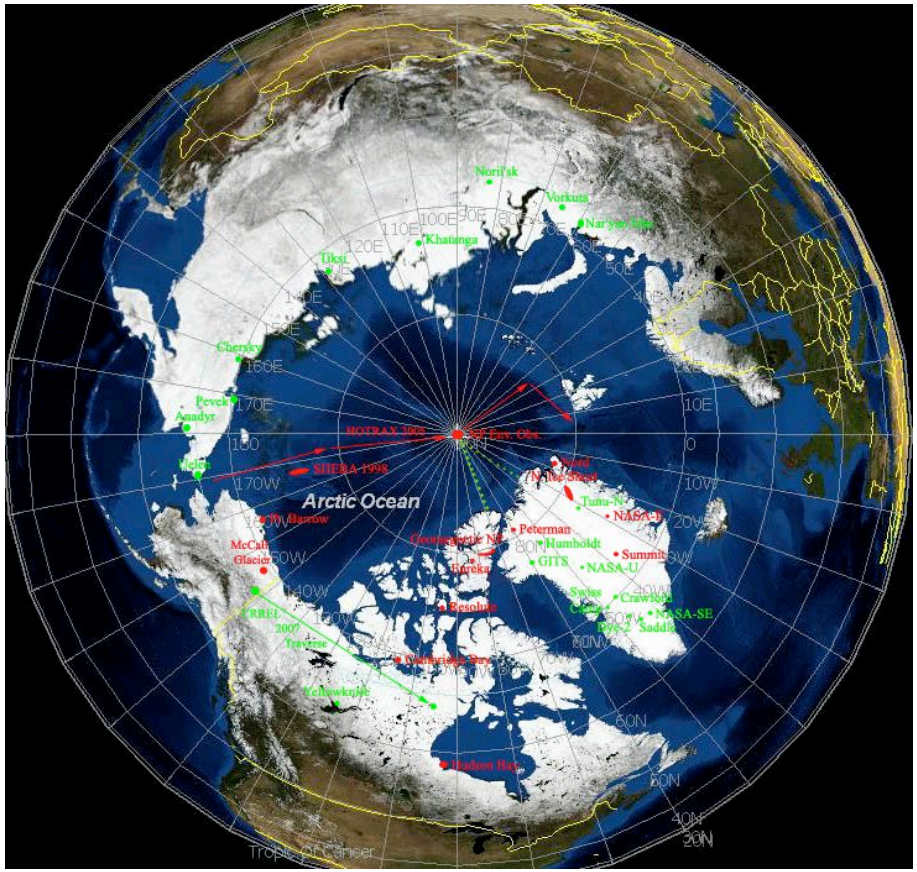
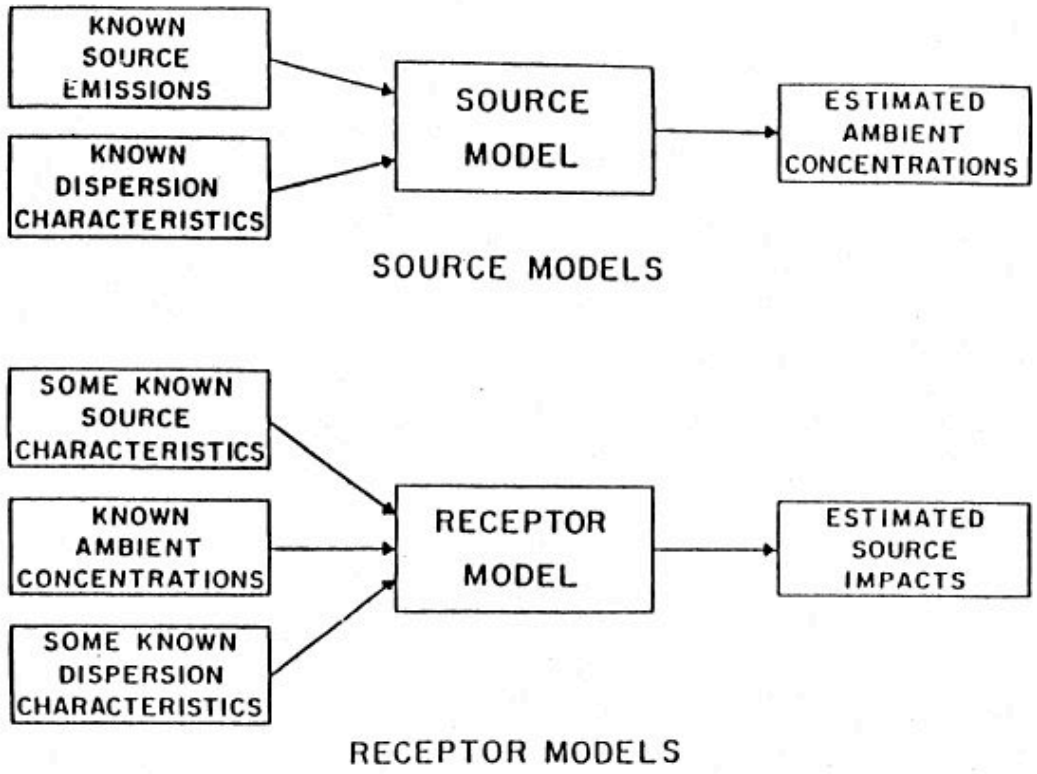
Black Carbon Source Attribution from Collected Samples



Source Model versus Receptor Model
A Basic Schematic

- The source model uses source emissions as inputs and calculates ambient concentrations
- The receptor model uses ambient concentrations as inputs and calculates source contributions



Adapted from Watson, 1979, *Dissertation*; Watson and Chow 2005

Receptor Models

- Source profiles "known"
  - Chemical Mass Balance
  - Other multivariate calibration models (Partial Least Squares, Artificial Neural Networks, Genetic Algorithms; for details see Seigneur and Hopke report available at: www.arb.ca.gov/airways/Modeling/References.htm)
- Source profiles unknown (ANOVA Techniques)
  - Absolute Principal Component Scores
  - UNMIX
  - Positive Matrix Factorization
  - Advanced Multivariate Models (e.g. Multi-linear Engine)
  - Empirical Orthogonal Function Analysis

How Multivariate Receptor Models Work
Synopsis

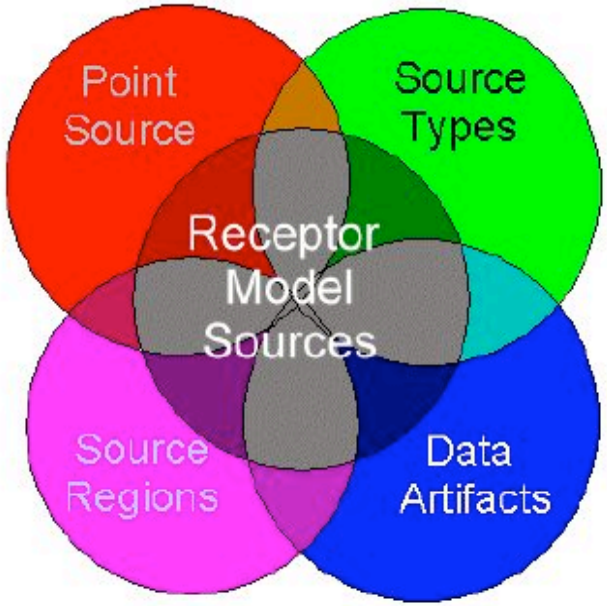
- Receptor models require the input of data of multiple species for multiple time periods and extract information from all sample data simultaneously. The reward for the extra complexity of these models is that they purport to estimate not only the source contributions but the source compositions (profiles) as well.
- Strengths
  - Use real ambient data to drive the model
  - Quantify sources in every sample
  - Give goodness-of-fit diagnostics for a robust analysis of how well the identified sources represent the data
- Weaknesses
  - Sources need to be independent to be isolated
  - Models rely on ambient data and their uncertainties
  - Transported and local sources may be difficult to isolate
  - Meteorology can obscure source signatures

Multivariate analyses essentially identify sources of covariation. We assume that the covariation comes from co-emission from a source, but it can also come from atmospheric processes that including meteorology and atmospheric chemistry. That is why we see OC with secondary sulfate and nitrate and also need an extra factor to explain the S-Se relationships in the eastern US.

What is a "Source"?

Source vs. Factor

- Receptor models identify factors that influence a data set
- These factors need to be related to actual source types or regions
- However, in addition to real sources, data artifacts and atmospheric chemistry influence the identification of factors
- Users must scrutinize results to ensure that influence from data artifacts is minimized, and that atmospheric chemistry effects (i.e., secondary production) are understood in the factors



From Poirot, 2001,
http://www.epa.gov/ttn/amtic/files/ambient/pm25/workshop/mdler.pdf

Natural Constraints

1. The original data must be reproduced by the model; *the model must explain the observations*
2. The predicted source compositions must be non-negative; *a source cannot have a negative percentage of an element*
3. The predicted source contributions to the aerosol must all be non-negative; *a source cannot emit negative mass*
4. The sum of the predicted elemental mass contributions from each source must be less than or equal to the total measured mass for each element; *the whole is greater than or equal to the sum of its parts*

Examples of EC Receptor Analysis from the Arctic

- Pollisar et al (JGR, 1998): PMF analysis for sources of Alaskan aerosol.
- Masclet et al (Atmos. Environ., 2000): UNMIX and PMF analysis on limited Greenland superficial snow samples.

Pollissar et al Sampling sites

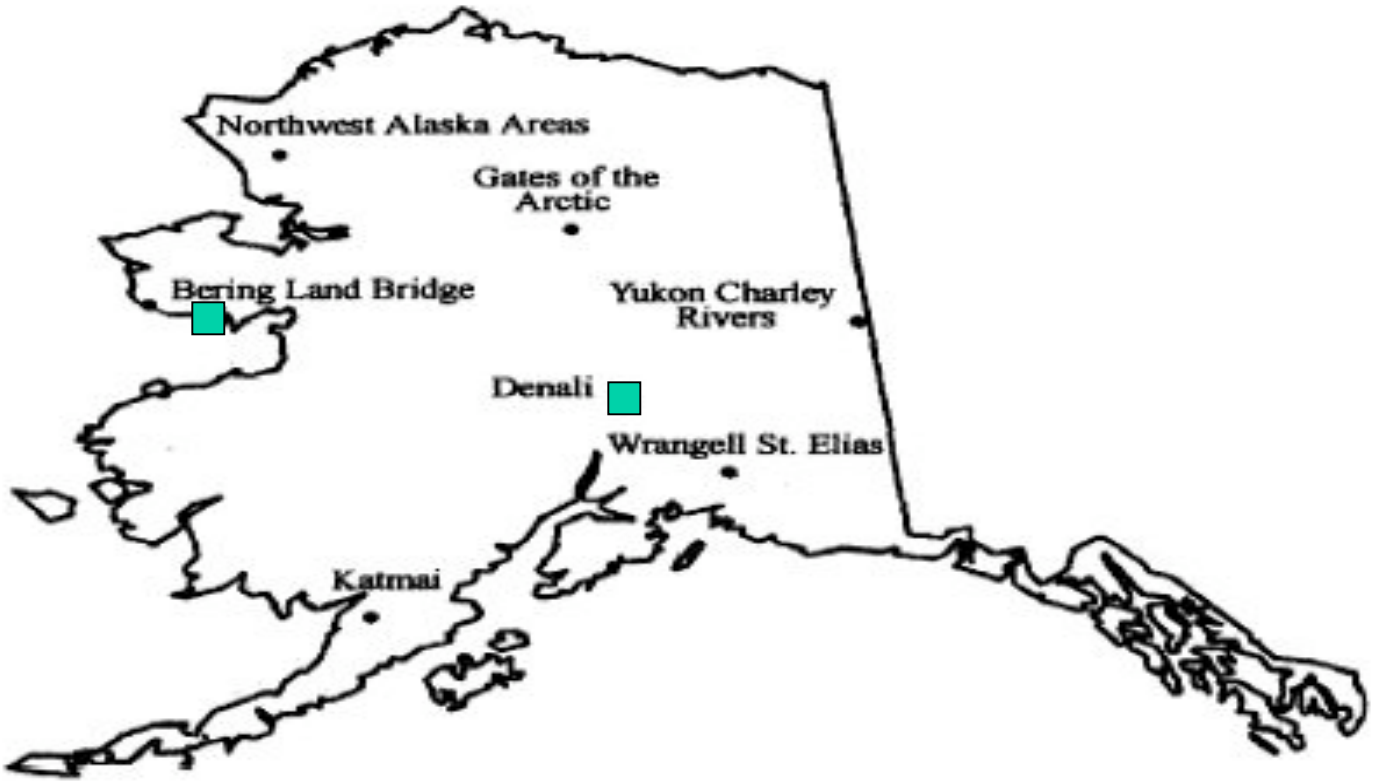
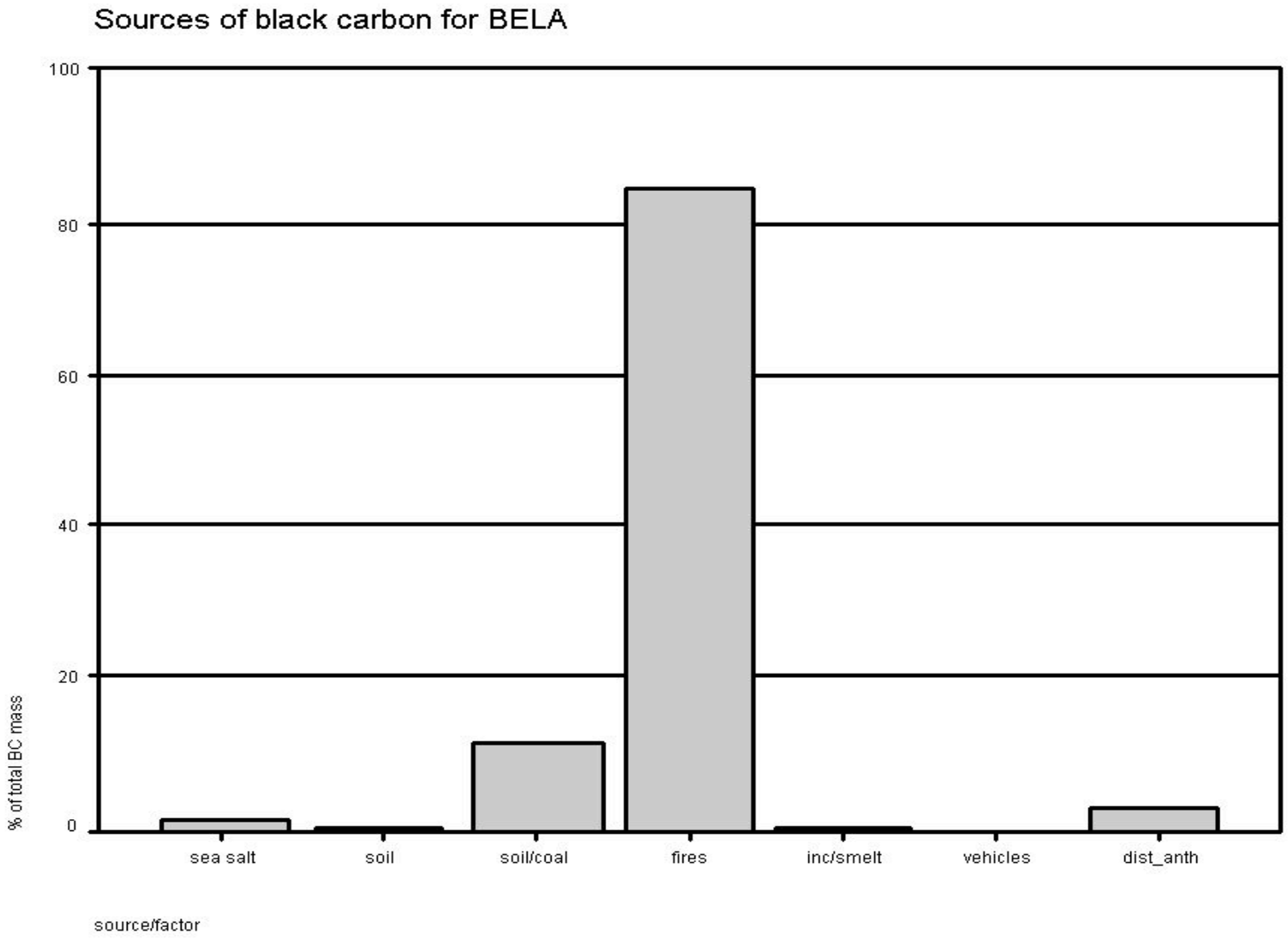
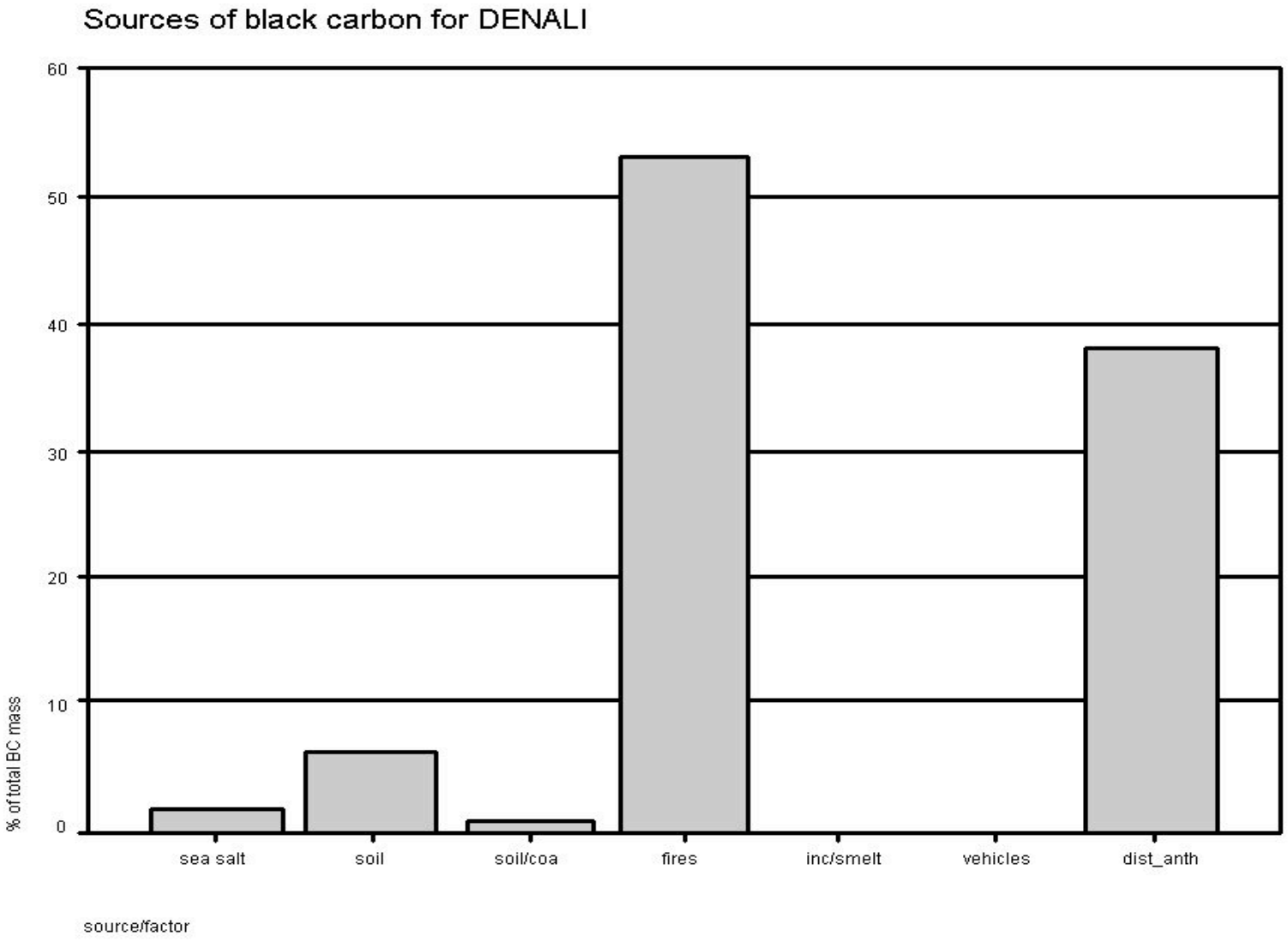
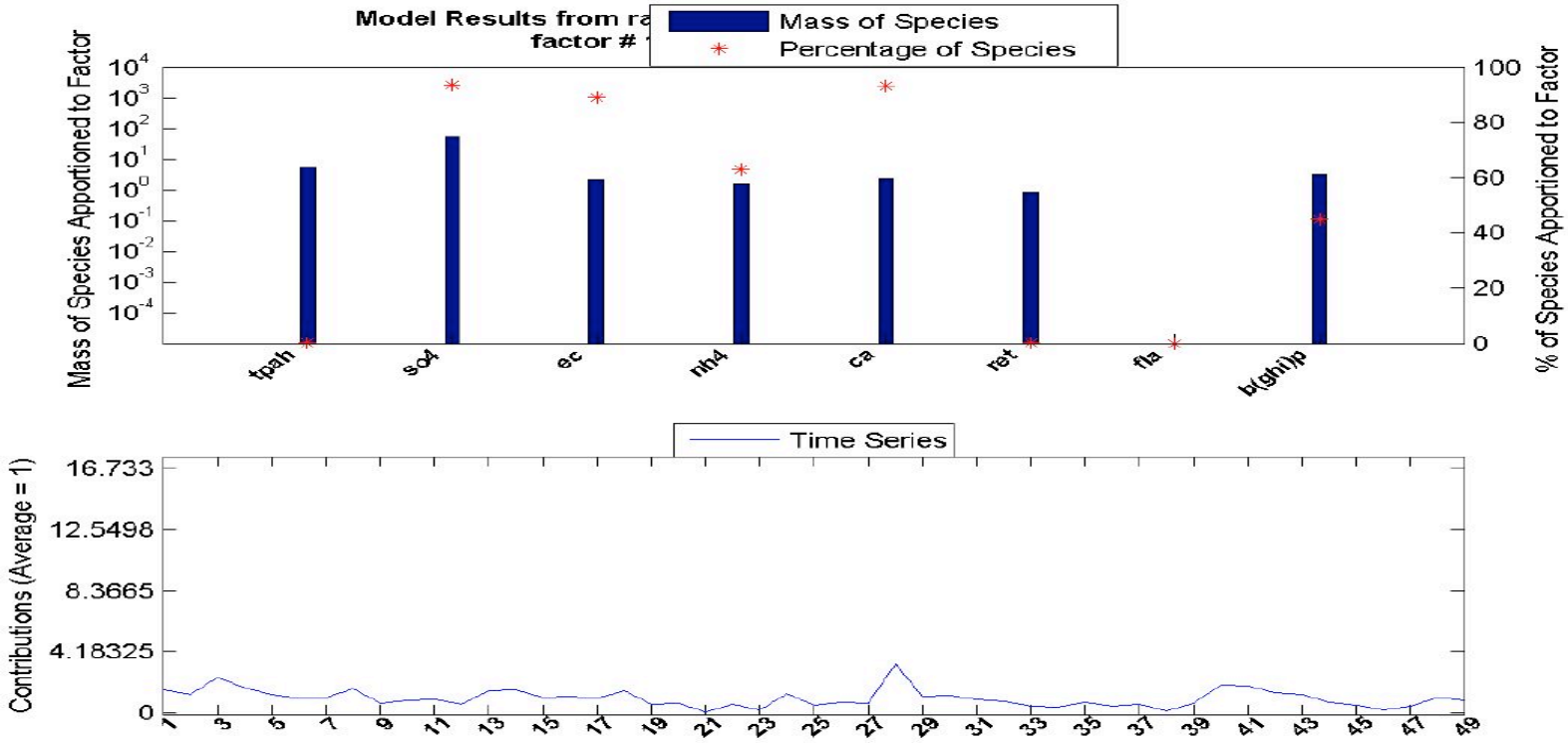


Figure 1. Locations of sites in Alaska National Park Service aerosol sampling network.

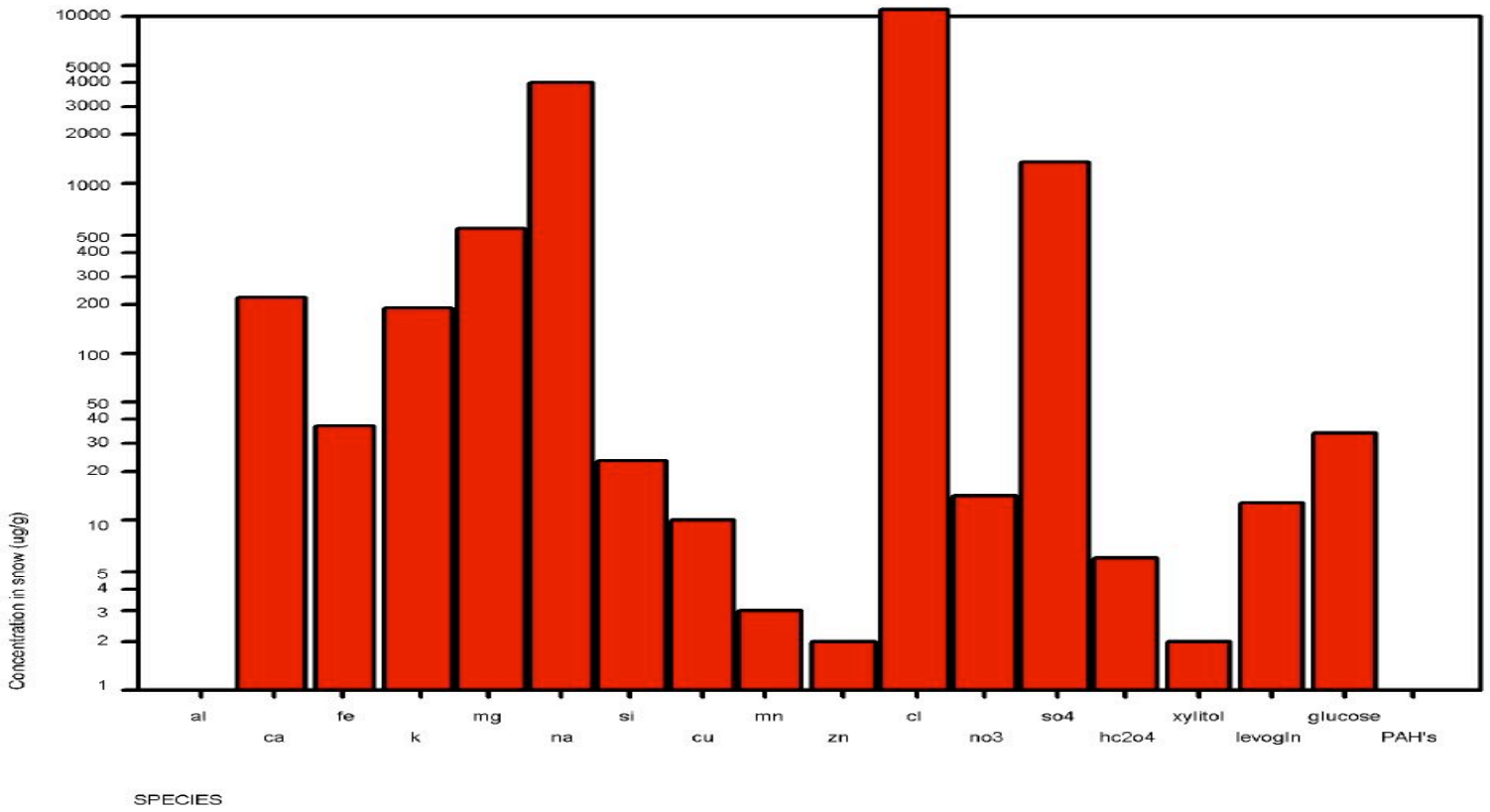

Sources of black carbon for BELA
100
80
60
40
20
0
% of total BC mass
sea salt
soil
soil/coal
fires
inc/smelt
vehicles
dist_anth
source/factor


Sources of black carbon for DENALI
60
50
40
30
20
10
0
% of total BC mass
sea salt
soil
soil/coa
fires
inc/smelt
vehicles
dist_anth
source/factor

Results of PMF analysis of Masclet et al (Summit, Greenland snow samples)



Analysis of the NPEO sample



Conclusions

- ANOVA receptor modeling is, in principle, a viable technique for snow EC source attribution in the arctic
- Interpretation of the factors as sources will require, as usual, SOME information on possible source profiles
- The suite of possible analytes in the filter samples is adequate but could be improved